IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of:

M. Matsumoto et al.   Group Art Unit: 4131

Serial Number: 10/583,506   Examiner: Brieann R. Fink

Filed: June 16, 2006

For: CONTACT LENS MATERIAL

DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

Sir:

Masahiro Matsumoto, citizen of Japan, duly deposes and says:

1. That he has graduated from Faculty of technology of Gifu University, Japan, in the year of 1990;

2. That he has been employed in his capacity since 1990 by MENICON CO., Ltd.;

3. That he has been engaged in research and development on a contact lens material;

DISCLOSURE OF INVENTION

The copolymer to be the CL material of the present invention can be obtained by polymerizing the mixture of the monomer components and the crosslinking agent at 40 to 120.degree. C. for 30 minutes to 5 hours. This is because excellent polymerization can be attained in a short time by substantially selecting and using the monomer components having excellent copolymerizability.

It is preferable that a polymerization temperature is suitably adjusted within the range from 40 to 120.degree. C. and is risen in the middle of polymerization. Since the polymerization is not completed or it takes a long time to polymerize in the polymerization reaction at a constant temperature in the low temperature range of about 40 to about 50.degree. C., it is required to raise the temperature. For example, when the initial temperature is set at 40 to 60.degree. C., the preliminary polymerization is performed at this temperature for 1 to 4 hours, then the temperature is then raised at 80 to 120.degree. C. to heat for 10 to 60 minutes, and the polymerization reaction can be completed. When the preliminary polymerization is not performed and the polymerization reaction is performed at least 70.degree. C. from the start, a polymerization time is preferably within 3 hours. In the case of the preliminary polymerization at the initial temperature of at most 40.degree. C., the polymerization reaction proceeds slowly, and it tends to take a long time to complete the polymerization reaction. At the polymerization temperature is more than 120.degree. C., the polymerization reaction goes out of control, and unpolymerized monomers tends to be increased.

The polymerization time is preferably 30 minutes to 5 hours. When the polymerization time is less than 30 minutes, the polymerization tends to be incomplete (the unpolymerized monomers are

increased). When the polymerization time is longer than 5 hours, the manufacturing time becomes longer, and as a result, the cost thereof tends to be high.

EXPERIMENTAL

Example 1

45 parts by weight of tris(trimethylsiloxy)silylstyrene (hereinafter, referred to as TMSiSt), 45 parts by weight of trifluoroethyl methacrylate (hereinafter, referred to as 3FEMA), and 10 parts by weight of ethylene glycol dimethacrylate (hereinafter, referred to as EDMA) were mixed. To 100 parts by weight of the obtained mixture, 0.3 part by weight of 2,2'-azobis(2,4-dimethylvaleronitrile) (hereinafter, referred to as V-65) is added to the obtained mixture as a polymerization initiator based on 100 parts by weight of this mixture, and the mixture was well stirred to be blended.

The mixture was injected into a mold made of polypropylene (diameter: 12 mm, depth: 5 mm), and the mold was covered with a PET (polyethylene terephthalate) film and sealed. After the mixture was preliminarily polymerized in a circulation drier charging with a nitrogen atmosphere at 50.degree. C. for 2 hours, the temperature was raised at 90.degree. C., and the mixture was kept with heating for t for 30 minutes to complete the polymerization (polymerization condition A).

The obtained copolymer in a state of a button shape was processed with cutting to prepare test pieces for measuring various properties. The properties of the test pieces were measured according to the following methods. Results are shown in Table 1.

Examples 2 to 9

Copolymers were prepared in the same manner as Example 1

except for changing the compositions thereof as shown in Table 1, and processed with cutting, and the properties of the obtained test pieces were measured according to the following methods. Results are shown in Table 1.

Comparative Examples 1 to 2 and 4 to 5

Copolymers were prepared in the same manner as Example 1 except for changing the compositions thereof as shown in Table 1, and processed with cutting, and the properties of the obtained test pieces were measured according to the following methods. Results are shown in Table 1.

Comparative Example 3

The copolymerizing components compounded according to Table 1 was injected into a glass test tube, after replacing a gas in the test tube with nitrogen, the test tube was sealed and put into a circulation thermostatic water bath. After the copolymerizing components were preliminarily polymerized at 35.degree. C. for 40 hours, a temperature was raised at 50.degree. C., and kept with heating for 8 hours. Then, in a circulation type thermostatic dryer, the copolymerizing components were heated at a speed of increasing the temperature of 10.degree. C./1.5 hours from 50 to 120.degree. C. to complete the polymerization (polymerization condition B).

The obtained copolymer was processed with cutting to prepare test pieces for measuring various properties. The properties of the test pieces were measured according to the following methods. Results are shown in Table 1.

Measurement of Properties
<Oxygen Permeability Coefficient (Dk)>

An oxygen permeability coefficient is measured at a temperature of 35.degree. C. for a measurement time of 2 minutes by using GTG(GAS to GAS) ANALYZER (manufactured by REHDERDEVELOPMENT COMPANY (U.S.)), and the obtained measured value is calculated by using Menicon EX (Dk value=64) standardized in ISO 9912-2 to find a Dk value. The results are shown in Table 1. The Dk value indicates a value $[(cm.sup.2/sec)(mLO.sub.2/(mLmmHg))]$ of an oxygen permeability coefficient, and a numerical value obtained by particularly multiplying the value of the oxygen permeability coefficient by 10.sup.-11.

<Water absorption ratio>

A set of ten test pieces (dried ones) having a thickness of 0.5 mm was prepared, and the weight (A [g]) was measured for every set, after immersing the set in a sample bottle containing distilled water at a temperature of 25.degree. C. for 24 hours, the wet weight (B [g]) of each test pieces is measured. A water absorption ratio is calculated by the following formula. The results are shown in Table 1. Water absorption ratio (%)={(B-A)/A}.times.100

<Remaining Component>

Referring to FDA GUIDANCE DOCUMENT FOR CLASS III CONTACT LENSES APRIL 1989 p.18 "Leachable and residual monomers", one plate having a diameter of 12 mm and a thickness of 0.5 mm is extracted by 5 mL of acetonitrile at 50.degree. C. for 72 hours. By using a high-speed liquid chromatography (HPLC; high-speed liquid chromatograph: 2695 separation module manufactured by WATERS Ltd.; detector: Photodiode Array Detector 996 manufactured by WATERS Ltd.; column: Develosil ODS HG-5 Length 250 mm.times.I.D. 4.6 mm, manufactured by Nomura Chemical Co., Ltd.), the concentration of each unpolymerized monomer component in the acetonitrile extraction solution is determined by the analytical curve produced from a standard

solution of each copolymerizing component, and the residual amount of the unpolymerized monomer component is calculated by the following formula. In the analysis condition of HPLC, the initial set value of a mobile phase is set to be acetonitrile/distilled water=30/70, and the composition of a mobile phase is continuously changed at the same time as initiation of the measurement (linear gradient), and the set value after 30 minutes is set to be acetonitrile/distilled water=100/0. The set value was maintained at acetonitrile/distilled water=100/0 until the analysis is terminated. A detection wavelength, a flow rate, an oven temperature and an amount to be charged are respectively set to be 210 nm, 1.0 mL/minute, 40.degree. C. and 20 .mu.L.

Residual amounts (% by weight) of each unpolymerized monomer component=concentration of each unpolymerized monomer component in acetonitrile extraction solution (10.sup.-6 g/mL).times.5(mL)/plate weight (g)).times.100

<Hardness>

Rockwell Superficial Hardness tester manufactured by Akashi Seisakusho Co., Ltd. is used, and the test pieces having a diameter of 12.0 mm and a thickness of 4.0 mm are measured by a load of 30 kg and 1/4 inches (about 0.64 cm) of an indenter in a thermohygrostat room with a temperature of 25.degree. C. and a relative humidity of 50%.

<Processability>

A test piece having a CL shape was processed by using a cutting machine using a diamond tool. Processability and the surface state of the sample piece after the process are visually observed, and evaluated based on the following evaluation criteria.

(Evaluation criteria)

largecircle.: The lens can be easily processed and Polished

DELTA.: The lens is processable, but a slight scuffing is

caused

X: Scuffing like chipping is significant, and the lens is difficult to be processed

<Flaw>

After polishing both faces of the obtained test piece and rubbing the faces with cloth, tissue paper or the like, the surface state of the test piece is visually observed, and evaluated based on the following evaluation criteria.

(Evaluation criteria)

largecircle.: Flaw is hardly observed [0264] .

DELTA.: Some flaws are observed

X: A lot of flaws are observed (even if the surfaces are rubbed with weak force, the surfaces are easily damaged).

<Shape Stability>

The polymerized copolymer in the shape of a button is processed with cutting, and a lens having a prescribed standard (base curve: 7.90 mm, power: -10.0 D (diopter), lens diameter: 9.2 mm) is prepared. After the lens was stored under a severe condition of 40.degree. C. in a wet state (surfactant-containing CL care article) and in a dry state for six months, the base curve and the lens diameter were measured.

All the lenses obtained from the copolymers of Examples had fluctuation of the base curve and lens diameter of within 0.05 mm both in both the wet state and in the dry state.

<Overall Evaluation>

In the polymerization condition A in which the total polymerization time is within 3 hours, a copolymer having the total remaining components of at most 2% by weight, Dk of at least 150, and the processability and the flaw of .largecircle. is evaluated as .largecircle., particularly, the copolymer having Dk of at least 180 is evaluated as .circleincircle. and the other one is evaluated as X.

TABLE 1

Components evaluation items etc		Ex. 1	2	3	4	5	6	7	8
TMSiSt(part by weight)		45	50	55	65	70		55	45
BTMSiSt							60		
3FEMA(part by weight)		45	35	35	30	20	30		
5FPMA								35	
6FIPMA									45
MAA(part by weight)									
Crosslinking agent (part by weight)	EDMA	10	15	10	5		10	10	
	VBMA					10			10
Ultraviolet absorber (part by weight)			0.25	0.25					
Colorant(part by weight)				0.005					
Polymerization initiator (part by weight)		0.3	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Polymerization condition		A	A	A	A	A	A	A	A
Dk value (GTG)		152	170	198	230	276	167	235	215
Water absorption (%)		0.06	0.05	0.06	0.06	0.05	0.06	0.05	0.07
Remaining component (% by weight)	TMSiSt	0.09	0.26	0.35	1.3	1.7		0.01	0.002
	BTMSiSt						1.0		
	3FEMA	0.28	0.06	0.06	0.01	N.D	0.07		
	5FPMA							0.32	
	6FIPMA								0.39
	MAA								
	Crosslinking agent	N.D	N.D	N.D	N.D	N.D	N.D	N.D	N.D
Hardness (30x)		55	56	53	43	41	52	46	39
Processability		O	O	O	O	O	O	O	O
Flaw		O	O	O	O	O	O	O	O
Overall evaluation		O	O	◎	◎	◎	O	◎	◎

Components evaluation items etc		Ex. 9	Com. Ex. 1	2	3	4	5
TMSiSt(part by weight)		60	45	55	55	40	75
BTMSiSt							
3FEMA(part by weight)			40	30	30	50	15
5FPMA							
6FIPMA		30					
MAA(part by weight)			5	5	5		
Crosslinking agent (part by weight)	EDMA		10	10	10	10	
	VBMA	10					10
Ultraviolet absorber (part by weight)				0.25	0.25		
Colorant(part by weight)				0.005	0.005		
Polymerization initiator (part by weight)		0.3	0.3	0.3	0.3	0.3	0.3
Polymerization condition		A	A	A	B	A	A
Dk value (GTG)		272	136	172	170	108	291
Water absorption (%)		0.07	0.5	0.4	0.4	0.05	0.04
Remaining component (% by weight)	TMSiSt	0.66	1.1	3.8	2.3	0.12	2.6
	BTMSiSt						
	3FEMA		2.3	1.2	0.11	2.4	N.D
	5FPMA						
	6FIPMA	0.28					
	MAA		4.8	3.9	2.2		
	Crosslinking agent	N.D	0.01	0.02	N.D	N.D	N.D
Hardness (30x)		34	39	22	48	51	27
Processability		O	O	Δ	O	O	X
Flaw		O	O	X	Δ	O	X
Overall evaluation		◎	X	X	X	X	X

TMSiSt: tris(trimethylsiloxy) silylstyrene
3FEMA: trifluoroethyl methacrylate
6FIPMA: hexafluoroisopropyl methacrylate
EDMA: ethylene glycol dimethacrylate
Ultraviolet absorber: 2-(5-chloro-2H-benzotriazole-2-yl)-6-(1,1-dimethylethyl)-4-methylphenol
Colorant: D&C; Green No. 6
N.D: less than detection limitation
BTMSiSt: bis(trimethylsiloxy)methylsilylstyrene
5FPMA: 2,2,3,3,3-pentafluoropropyl methacrylate
MAA: methacrylic acid
VBMA: 4-vinylbenzyl methacrylate
Polymerization initiator: 2,2'-azobis(2,4-dimethylvaleronitrile)

The undersigned declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This 16th day of January, 2009

by *Masahiro Matsumoto*

Masahiro Matsumoto

We, the undersigned witnesses, hereby acknowledge that Masahiro Matsumoto is personally known to us and did execute the foregoing Declaration in our presence on:

Date: January 16, 2009   Witness *Ryuzochi Watanabe*

Date: January 16, 2009   Witness *Kunihisa Yoshida*